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Upper critical field measurements up to 60 T in arsenic-deficient $\text{LaO}_{0.9}\text{F}_{0.1}\text{FeAs}_{1-\delta}$: Pauli limiting behaviour at high fields vs. improved superconductivity at low fields

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Abstract We report resistivity and upper critical field $B_{c2}(T)$ data for As deficient $\text{LaO}_{0.9}\text{F}_{0.1}\text{FeAs}_{1-\delta}$ in a wide temperature and high field range up to 60 T. These disordered samples exhibit a slightly enhanced superconducting transition at $T_c = 29$ K and a significantly enlarged slope $dB_{c2}/dT = -5.4$ T/K near T_c which contrasts with a flattening of $B_{c2}(T)$ starting near 23 K above 30 T. This flattening is interpreted as Pauli limiting behaviour (PLB) with $B_{c2}(0) \approx 63$ T. We compare our results with $B_{c2}(T)$ -data reported in the literature for clean and disordered samples. Whereas clean samples show no PLB for fields below 60 to 70 T, the hitherto unexplained flattening of $B_{c2}(T)$ for applied fields $H \parallel ab$ observed for several disordered closely related systems is interpreted also as a manifestation of PLB. Consequences of our results are discussed in terms of disorder effects within the frame of conventional and unconventional superconductivity.

Keywords pnictide superconductors, upper critical field

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The recently discovered FeAs based superconductors¹ exhibit high transition temperatures $T_c \leq 57$ K and remarkably high upper critical fields $B_{c2}(0)$ exceeding often 70 T. Many basic properties of these novel superconductors and the underlying pairing mechanism are still not well understood. A study of $B_{c2}(T)$, in particular, investigations on disordered FeAs superconductors are of large interest since for an unconventional pairing both T_c and dB_{c2}/dT at T_c are expected to be

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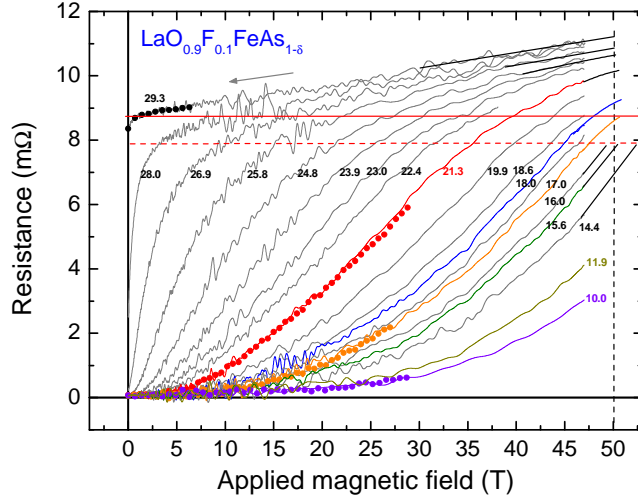


Fig. 1 (Color online) Field dependence of the resistance at fixed T (see legend) measured in pulsed fields. Lines: measurements up to 47 T; symbols measurements up to 29 T shown for selected T . Horizontal full and dashed lines: $R = R_N$ and $R = 0.9R_N$, respectively with R_N as the resistance in the normal state.

suppressed by introducing disorder. In the present paper, $B_{c2}(T)$ of As-deficient $\text{LaO}_{0.9}\text{F}_{0.1}\text{FeAs}_{1-\delta}$ samples is studied in fields up to 60 T.

Polycrystalline samples of $\text{LaO}_{0.9}\text{F}_{0.1}\text{FeAs}$ were prepared by the standard solid state reaction method^{2,3}. Some samples have been wrapped in a Ta foil during the final annealing procedure. Ta acts as an As getter at high temperatures forming a solid solution of about 9.5 at.% As in Ta. This leads to an As loss in the samples resulting in an As/Fe ratio of about 0.9. Due to disorder in the FeAs layer, an enhanced resistivity in the normal state at 31 K is found for the investigated As-deficient sample (ADS) exceeding that of a clean reference sample by a factor of about three. Nevertheless, the ADS has, with $T_c = 28.5$ K, a higher T_c than stoichiometric reference samples ($T_c = 27.7$ K)³.

In Fig. 1, resistance data obtained in pulsed fields up to 50 T are shown for the ADS. Gold contacts (100 nm thick) were prepared by sputtering in order to provide a low contact resistivity and, therefore, to avoid possible heating effects. The magnetic field generated by the employed IFW's pulsed field magnet rises within 10 ms to its maximum value B_{max} and decreases afterwards to zero within the same time. The resistance data shown in Fig. 1 were taken for descending field using $B_{max} = 47$ T. Additionally, resistance data were collected for $B_{max} = 29$ T at several selected temperatures. The agreement between the data obtained for both B_{max} -values confirms that our data are not affected by sample heating.

For polycrystalline samples, only the highest upper critical field B_{c2}^{ab} is accessible which is related to those grains oriented with their ab -planes along the applied field. B_{c2}^{ab} was determined from the onset of superconductivity (SC) defining it at 90% of the resistance R_N in the normal state (see Fig. 1). The temperature dependence of B_{c2}^{ab} of our As-deficient sample obtained from pulsed field measurements in the IFW and the FZD is shown in Fig. 2 together with B_{c2} data

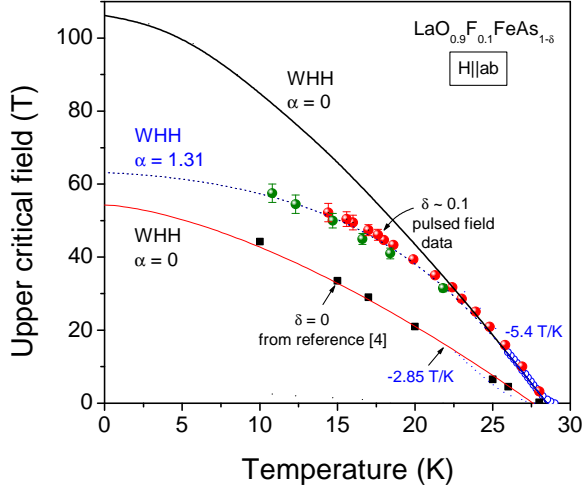


Fig. 2 (Color online) T -dependence of B_{c2}^{ab} . Data for the As-deficient sample from DC (■) and pulsed field measurements (● - IFW Dresden, ● - FZD) and data for a clean reference sample⁴. Solid lines: WHH model without PLB. Dotted line: $B_{c2}^*(T)$ for $\alpha = 1.31$ (see text).

reported for a clean reference sample⁴. The large slope $dB_{c2}/dT = -5.4$ T/K at T_c of our ADS points to strong impurity scattering in accord with its enhanced resistivity at 30 K. For the clean sample⁴ the available data up to 45 T is well described by the WHH (Werthamer-Helfand-Hohenberg) model⁵ for the orbital limited upper critical field. Whereas for the ADS, the WHH model which predicts $B_{c2}^*(0) = 0.69T_c(dB_{c2}/dT)|_{T_c} = 106$ T at $T = 0$, fits the experimental data up to 30 T, only. For applied fields above 30 T increasing deviations from the WHH curve are clearly visible both for the $B_{c2}(T)$ data from the IFW and the FZD. The flattening of $B_{c2}(T)$ at high field points to its limitation by the Pauli spin paramagnetism. This effect is measured in the WHH model by the Maki parameter $\alpha = \sqrt{2}B_{c2}^*(0)/B_p(0)$, where $B_p(0)$ is the Pauli limiting field. The paramagnetically limited upper critical field, B_{c2}^p , is given by $B_{c2}^p(0) = B_{c2}^*(0)(1 + \alpha^2)^{-0.5}$. For our ADS, a satisfying fit of the experimental data to this model was obtained for $\alpha = 1.31$ (see Fig. 2) and yields $B_{c2}^p(0) = 63$ T.

For several disordered closely related systems, a similar flattening of $B_{c2}(T)$ as we found for our ADS has been reported for applied fields $H \parallel ab$. This is shown in Fig. 3 where the normalized upper critical field $h^* = B_{c2}(T)/[T_c(dB_{c2}/dT)|_{T_c}]$ is plotted against the reduced temperature $t = T/T_c$. In contrast, $B_{c2}(T)$ data for clean $\text{LaO}_{0.93}\text{F}_{0.07}\text{FeAs}$ samples⁶ (see Fig. 3) show almost no Pauli-limiting behavior for fields up to 70 T. The data in Fig. 3 are well described by the WHH model using the obtained Maki parameters α . The deviation of $h^*(t)$ at low T from $h^*(t)$ for $\alpha = 0$ increases with α due to rising paramagnetic pair-breaking.

We found for our As-deficient samples indications for a strongly enhanced Pauli paramagnetism from μSR experiments¹¹. Their improved SC at high T and low fields can be understood within conventional s-wave SC by enhanced disorder. In contrast, for clean FeAs superconductors an unconventional s^\pm -wave sce-

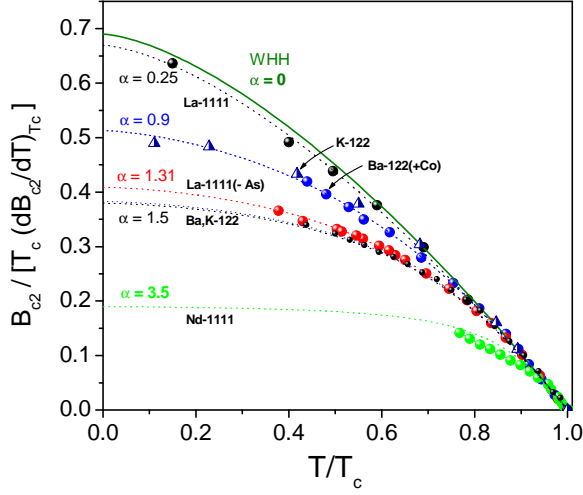


Fig. 3 (Color online) Normalized upper critical field $B_{c2}(T)/[T_c (dB_{c2}/dT)|_{T_c}]$ vs. T/T_c for an As-deficient $\text{LaO}_{0.9}\text{F}_{0.1}\text{FeAs}_{1-\delta}$ sample (La-1111(-As)) in comparison with data reported for non-deficient $\text{LaO}_{0.93}\text{F}_{0.07}\text{FeAs}$ (La-1111, $T_c = 25$ K)⁶, $\text{Ba}(\text{Fe}_{0.9}\text{Co}_{0.1})_2\text{As}_2$ (Ba-122(+Co), $T_c = 21.9$ K)⁷, KFe_2As_2 (K-122, $T_c = 2.8$ K)⁸, $\text{Ba}_{0.55}\text{K}_{0.45}\text{Fe}_2\text{As}_2$ (Ba-122, $T_c = 32$ K)⁹, $\text{NdO}_{0.7}\text{F}_{0.3}\text{FeAs}$ (Nd-1111, $T_c = 45.6$ K)¹⁰. Dotted and solid lines: WHH model for the indicated values. All curves shown correspond to $H \parallel ab$.

nario has been proposed. On the basis of our results for $B_{c2}(T)$, two alternative scenarios of opposite disorder effects might be suggested: (i) an impurity-driven change of the pairing state from s^\pm to conventional s_{++} -wave SC and (ii) a special impurity-driven stabilization of the s^\pm state where the As-vacancies are assumed to scatter predominantly within the bands. The PLB found here suggests to continue measurements at least up to 70 T in order to elucidate, whether there is still much room for increasing B_{c2} beyond that range. The possibility to improve the low-field properties of FeAs superconductors by introducing As vacancies opens new preparation routes for optimising the properties of these superconductors.

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